Improved quantification of Chinese carbon fluxes using CO_2/CO correlations in Asian outflow

Parvadha Suntharalingam[1], Daniel J. Jacob, Paul I. Palmer, Jennifer A. Logan, Robert M. Yantosca, Yaping Xiao, Mathew J. Evans,
Division of Engineering and Applied Sciences, Harvard University, Cambridge, MA

David G. Streets,
Argonne National Laboratory, Argonne, IL

Stephanie L. Vay, Glen W. Sachse
NASA Langley Research Center, VA

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[1] Corresponding author:
Parvadha Suntharalingam, Division of Engineering and Applied Sciences, Harvard University, Pierce Hall, 29, Oxford St., Cambridge, MA 02138
pns@io.harvard.edu

Abstract.

We use observed CO_2:CO correlations in Asian outflow from the TRACE-P aircraft campaign (February-April, 2001), together with a 3-D global chemical transport model (GEOS-CHEM), to constrain specific components of the East Asian CO_2 budget including in particular Chinese emissions. The CO_2/CO emission ratio varies with the source of CO_2 (different combustion types vs. the terrestrial biosphere) and provides a characteristic signature of source regions and source type. Observed CO_2/CO correlation slopes in East Asian boundary layer outflow display distinct regional signatures ranging from 10-20 mol/mol (outflow from northeast China) to 80 mol/mol (over Japan). Model simulations using best a priori estimates of regional CO_2 and CO sources from Streets et al. [2003] (anthropogenic), the CASA model (biogenic), and Duncan et al. [2003a] (biomass burning) overestimate CO_2 concentrations and CO_2/CO slopes in the boundary layer outflow. Constraints from the CO_2/CO slopes indicate that this must arise from an overestimate of the modeled regional net biospheric CO_2 flux. Our corrected best estimate of the net biospheric source of CO_2 from China for March-April 2001 is 3200 Gg C/day, which represents a 45% reduction of the net flux from the CASA model. Previous analyses of the TRACE-P data had found that anthropogenic Chinese CO emissions must be about 50% higher than in the Streets et al. [2003] inventory. We find that such an adjustment improves the simulation of the CO_2/CO slopes and that it likely represents both an underreporting of sector activity (domestic and industrial combustion) and an underestimate of CO emission factors. Increases in sector activity would imply increases in Chinese anthropogenic CO_2 emissions and would also imply a further reduction of the Chinese biospheric CO_2 source to reconcile simulated and observed CO_2 concentrations.

1. Introduction

Inverse model analyses have emerged as an important tool for improving our understanding of the carbon cycle. These analyses have employed atmospheric observations of CO_2 alone [Bousquet et al. 1999; Fan et al. 1998; Rayner et al. 1999], as well as O_2/N_2 ratios [Keeling et al. 1996; Manning et al. 2001; Bender et al. 1996] and δC^{13} [Francey et al.1999] to place constraints on global and regional scale carbon exchanges with the atmosphere. They concur on such large-scale features as the existence of significant net terrestrial carbon uptake at northern mid-latitudes in the late 1980s and early 1990s [Bousquet et al. 2000, 1999; Fan et al. 1998]. However, they are limited by inadequate observational network coverage [Gloor et al. 2000; Suntharalingam et al. 2003] and by transport model errors [Gurney et al. 2002; Peylin et al. 2002].

Atmospheric CO_2 shares common combustion sources with a number of trace gases, in particular CO. The CO_2/CO emission ratio varies with the efficiency of combustion and thus provides a characteristic signature of source regions and source type. High resolution measurements of CO_2 and CO are available from a number of aircraft missions but have yet to be exploited for their information on carbon fluxes. In this study we use the variable CO_2:CO correlations in Asian outflow measured on the TRACE-P aircraft mission [Jacob et al., 2003], together with a global 3-D model, to explore the constraints that they provide on the combustion sources of CO_2 and to separate these contributions from biospheric CO_2 fluxes.

Use of CO to identify combustion and biospheric sources of CO_2 has been demonstrated previously. Potosnak et al. [1999], using continuous surface air measurements at Harvard Forest in Massachusetts, employed CO as a predictor of CO_2 emissions from fossil fuel burning in a study of contributions to observed CO_2 variability. A recent analysis by Lagenfelds et al. [2002] identifies the role of biomass burning in inter-annual atmospheric CO_2 variations by using flask samples of CO_2, CO, δC^{13} and other species from the CSIRO observational network. These studies used isolated surface observations. As we will show here, aircraft observations of CO_2 and CO in continental outflow provide robust information on the diversity of regional signatures.

The TRACE-P campaign, flown from bases in Hong Kong and Yokota (Japan), focused on the quantification of Asian pollution export to the Pacific [Jacob et al., 2003]. Although the aircraft could not fly over land, except for Japan, observations for a range of outflow regions offer a powerful resource for exploring the use of CO_2/CO correlations to constrain components of the Asian carbon budget. Gridded anthropogenic emission inventories of CO_2 and CO in eastern Asia were generated from socioeconomic data to serve as an a priori for the TRACE-P mission [Streets et al. 2003]. These inventories offer a context for interpreting the aircraft observations when implemented into a 3-D chemical transport model (CTM), as will be done here.

2. The TRACE-P campaign

Measurements in TRACE-P were collected by two research aircraft, a DC-8 and a P-3B, from February 24 to April 10, 2001. Asian outflow in early spring is particularly strong and is almost exclusively to the Pacific [Liu et al., 2003]. Considerable biomass burning takes place in Southeast Asia in that season and makes a major contribution to the outflow. A detailed overview of mission objectives, flight specifics, and results to date is provided in Jacob et al. [2003]. The DC-8 and P-3B had ceilings of 12 and 7 km respectively. A total of 38 flights were conducted, of which 13 were transit flights from the United States to the western Pacific. We focus here on the western Pacific data (Figure 1) collected from March 1 to April 6, 2001.

Descriptions of meteorological patterns and outflow pathways during TRACE-P are presented in Fuelberg et al. [2003] and Liu et al. [2003]. Export of Asian pollution to the Pacific was driven by wave cyclones and the associated cold fronts sweeping across East Asia along a northwest to southeast trajectory. The primary outflow pathways were (a) lifting to the free troposphere in the southwesterly Warm Conveyor Belts (WCBs) ahead of the fronts, and (b) boundary layer advection behind the fronts [Liu et al., 2003]. Lifting of Asian air by the WCBs took place over a wide range of latitudes from 15^{o} to 40^{o}N, resulting in considerable mixing of anthropogenic and biomass burning effluents in the free tropospheric outflow

[Carmichael et al. 2003; Ma et al. 2003; Miyazaki et al. 2003]. In contrast, boundary layer outflow was devoid of biomass burning influence [Liu et al. 2003]. The highest concentrations of CO_2, CO, and other species were measured on flights sampling this boundary layer outflow off the east coast of China [Vay et al., 2003; Carmichael et al, 2003; Bartlett et al., 2003; Simpson et al. 2003].

CO_2 measurements were made on both aircraft using non-dispersive infrared spectrometers (modified Li-Cor model 6252). Details of measurement procedures are provided in Anderson et al. [1996] and the TRACE-P data are presented in Vay et al. [2003]. CO was measured using a differential absorption spectroscopic method (DACOM, Sachse et al., [1987]). Analyses of the TRACE-P CO measurements are presented in a number of papers focused on quantifying Asian sources of CO [Carmichael et al. 2003; Palmer et al. 2003a] or using CO as a tracer of Asian outflow [Heald et al., 2003b; Russo et al., 2003; Kiley et al., 2003; Palmer et al., 2003b]. Our analysis of CO_2/CO correlations (section 4) uses 1-minute averages of the data, and our comparison to CTM simulations in section 6 uses data averaged over the CTM grid resolution (2^ox2.5^o in the horizontal and 48 vertical sigma levels).

3. A Priori Asian emission inventories

The early spring timing of the TRACE-P campaign coincided with the biomass burning season in Southeast Asia [Duncan et al. 2003a] and with the seasonal maximum in regional biospheric efflux. In that season, regional CO_2 emission from terrestrial biospheric respiration dominates over photosynthetic uptake. Figure 2 presents the seasonal variation of CO_2 surface fluxes for the East Asian region (aggregated over the ensemble of regions in Figure 3) based on a priori inventories presented below. The net source of CO_2 from East Asia is at a maximum in March-April and includes contributions of comparable magnitude from the terrestrial biosphere, fossil fuel combustion, and biomass burning.

The biomass burning contribution is often omitted in long-term analyses of atmospheric CO_2 data. Although it is a large source of CO_2 [Levine et al., 1994], it is largely balanced by vegetation regrowth over the next year [Houghton and Hackler, 1999]. Biomass burning does, however, provide an important seasonal source of CO_2

that must be included in analyses of short-term data such as those provided by aircraft campaigns.

Streets et al. [2003] developed gridded anthropogenic emission inventories for East Asia, with associated uncertainty estimates, in support of the TRACE-P mission. We rely on this study for the a priori sources of CO_2 and CO from fossil fuel and biofuel. Biomass burning emissions in February-April 2001 were close to the climatological average as indicated by the TOMS Aerosol Index [Heald et al. 2003a]. We use here a global February-April climatology of biomass burning emissions [Duncan et al., 2003a], distributed daily on the basis of satellite fire count data for the TRACE-P period [Heald et al. 2003a]. We also account for the small secondary sources of CO from oxidation of anthropogenic and biogenic non-methane hydrocarbons (NMHCs), which are computed as described by Duncan et al. [2003b].

CO_2 exchange with the terrestrial biosphere is based on monthly gridded fields of Net Primary Production (NPP) and heterotrophic respiration provided by the CASA ecosystem model [Potter et al., 1993; Field et al. 1995; Randerson et al. 1997]. The climatological CASA fluxes simulate the seasonal cycle of the balanced biosphere (i.e., fluxes at any location integrate to zero over the span of a year). This inventory does not account for CO_2 emissions from biomass burning (which we account for separately as outlined above) or for the subsequent carbon uptake by regrowth. The TRACE-P campaign was conducted before the start of the regional growing season and hence the contribution of this latter component should have little effect on our analysis.

Table 1 presents a breakdown of the a priori CO_2 emissions on a regional and source-specific basis for the TRACE-P measurement period. The regions considered are those of Figure 3. CO_2 emissions from China dominate the East Asian total (55%), and emissions from the biosphere represent 64% of the Chinese total. Biomass burning represents about half of the CO_2 source from Southeast Asia. Fossil fuel combustion is the principal source in Japan and Korea (68% and 66%, respectively). The a priori CO sources are also listed in Table 1. The main regional contributions to atmospheric CO are from Chinese fossil and biofuel combustion, and Southeast Asian biomass burning.

Table 2 lists the regional and source-specific CO_2/CO molar emissions ratios derived from these a priori inventories. Regional totals are of most interest as the continental outflow sampled by the aircraft integrates contributions from different processes within a source region. As seen from the table, the combustion sources are characterized by a range of ratios that reflect differences in combustion efficiency and in air pollution controls for CO. One can also define a CO_2/CO ratio for the biospheric source on the basis of CO production from short-lived biogenic NMHCs, mainly isoprene. This ratio is generally very high relative to that from combustion (Table 2). Southeast Asia is an exception with a low value of 11. In this tropical region, the opposing NPP and respiration fluxes of CO_2 are large and almost equal, yielding an NEP flux close to zero. The combination of low NEP CO_2 emissions and high production of CO from local biogenic NMHC emissions results in the low biospheric CO_2/CO ratio.

The overall CO_2/CO source signature of a region is then determined by the relative proportions of the different sources. In China, the low-efficiency biomass combustion sources display low CO_2/CO emission ratios of about 12 (mol/mol), whereas fossil fuel combustion has an emission ratio of 26. The overall ratio for China is 56, reflecting a major contribution from the terrestrial biosphere. In Japan, air pollution controls for CO result in a high fossil fuel ratio of 117. Contributions from biomass burning and biofuel use are relatively small there, as is the biospheric flux. This results in an overall Japanese CO_2/CO ratio of 122, a factor of two higher than for China. The lowest CO_2/CO emission ratios are for India and Southeast Asia, reflecting the major influences of biomass burning and biofuel combustion in these regions.

4. Observed CO_2/CO correlations and relationships to sources

We illustrate some of the distinct regional signatures seen in the TRACE-P CO_2/CO correlations by using data from two flights in conjunction with back-trajectory calculations from Fuelberg et al. [2003]. The CO_2/CO slopes in this analysis and in the remainder of the study are obtained using the reduced major axis method.

Flight DC8 #16, (March 29, 2001)

Figure 4 shows the CO_2/CO relationships observed on DC-8 flight 16. This was a sunrise flight that ascended out of Yokota Air Force Base, Tokyo, and made repeated measurements of the same vertical profile over the East China Sea (124^o-125^o E, 28.5^o N) before returning to Japan. The aircraft sampled a heavy Chinese pollution plume in the boundary layer, capped by clean subsiding air in the free troposphere above 2 km. The CO_2/CO slopes in Figure 4 show 3 distinct populations which we discuss in turn.

- Population 1 : Ascent out of Tokyo (surface to 3 km). The CO_2/CO slope of 65 mol/mol is at the high end of the range observed in the boundary layer during TRACE-P. Emission inventories indicate a Japanese CO_2/CO source signature of 122 (Table 2). The CO_2/CO slope measured here is about 40% lower. Other analyses of TRACE-P boundary layer measurements over Japan point to the influence of transport from Korean and Chinese sources upwind [Palmer et al., 2003b; Blake et al. 2003] and this could explain the discrepancy. We will return to this point later.
- Population 2 : Chinese boundary layer outflow. The CO_2/CO slope is only 12 mol/mol. Back trajectories indicate a dominant influence from the Shanghai region. The CO_2/CO slope is typical of biofuel or biomass burning and much lower than would be expected regionally for China (Table 2). Based on their analysis of TRACE-P flights over the East China and Yellow Seas, Carmichael et al. [2003] concluded that Chinese domestic fuel emissions for CO in the Streets et al. [2003] inventory are greatly underestimated. Such a source would be expected to have a low CO_2/CO emission ratio. Again, we will return to this point later.
- Population 3 : Background free tropospheric air. Concentrations are low and the CO_2/CO slope is 60 mol/mol. Correlation between CO_2 and CO would be expected to reflect the common large-scale latitudinal gradients of the two gases during that season. This is consistent with our global CTM simulation for the TRACE-P period (section 5), which indicates a mean CO_2/CO

correlation slope of 72 in free tropospheric background air upstream of East Asia.

Flight P3B #14 (March 18, 2001)

This flight sampled boundary layer outflow south of Japan and over the Yellow Sea, downwind of northern China and Korea. The outflow was capped by a strong subsidence inversion at 2 km altitude. As seen in Figure 5, the CO_2/CO correlations measured on this flight fall into distinct populations.

- Population 1: Boundary layer outflow sampled south of Japan. This population is characterized by a high CO_2/CO slope of 75. Back trajectories indicate boundary layer influences from the Chinese coastal region between Shanghai and Qingdao, as well from South Korea and the southern tip of Japan. Blake et al. [2003] find a strong Korean and Japanese influence on these flight legs as indicated by high levels of methyl bromide. This could explain the high CO_2/CO slope.
- Population 2: Chinese boundary layer outflow over the Yellow Sea. The CO_2/CO slope is 22. The CO measurements are some of the highest in the campaign. Back trajectories indicate surface influence from the Beijing/Tianjin region as well as further south towards Qingdao. Characteristics of these plumes are discussed in Weber et al. [2003] and Carmichael et al. [2003]. As with Population 2 from DC-8 flight 16, the low CO_2/CO slope could result from domestic combustion sources.
- Population 3 : Subsiding air at 2-3 km altitude. This population displays a high CO_2/CO slope (80) typical of the free tropospheric background discussed previously.

5. Global model simulation of CO_2 and CO for the TRACE-P period

Quantitative interpretation of the observed CO_2/CO slopes in terms of their constraints on sources requires a 3-D model to resolve the effects of transport and associated mixing of different source types and source regions. We simulate CO_2 and

CO with the GEOS-CHEM CTM (version 4.33) driven by assimilated meteorological observations from the Goddard Earth Observation System (GEOS) of the NASA Data Assimilation Office (DAO). The horizontal resolution is 2^{o}x2.5^{o} with 48 vertical sigma levels and a dynamic time step of 15 minutes. The original description of the GEOS-CHEM CTM is given by Bey et al. [2001]. Applications of the CO simulation for the TRACE-P period to constrain East Asian fossil fuel and biomass burning emissions are presented by Palmer et al. [2003a] and Heald et al. [2003b]. Further applications of GEOS-CHEM to the interpretation of CO data from TRACE-P are presented by Li et al. [2003], Liu et al. [2003] and Jaegle et al. [2003]. In an intercomparison of seven different global and regional models simulating CO for the TRACE-P period, Kiley et al. [2003] found no overall bias in the GEOS-CHEM simulation of transport. Based on statistics of the difference between aircraft measurements and model simulations, Palmer et al. [2003a] estimated a 20-30% transport error for the GEOS-CHEM simulation of CO in TRACE-P.

Simulation of CO_2 is a new GEOS-CHEM capability that draws on our previous CO_2 simulation with the GISS II' CTM [Suntharalingam et al. 2003]. We have evaluated the global characteristics of the GEOS-CHEM CO_2 simulation (surface distributions and detrended seasonal cycles) using data from the GLOBALVIEW-CO_2 network. The modeled distribution reproduces the observed large-scale spatial gradients, as well as the amplitude and phasing of the seasonal cycle at most of the GLOBALVIEW observation sites. For the TRACE-P simulations, we combine the regional Asian emission inventories of CO_2, discussed in section 3, with standard global inventories for the rest of the world in 2001. These include CO_2 fluxes from fossil fuel combustion [Marland et al. 2001], biofuel combustion [Yevich and Logan, 2003], biomass burning [Duncan et al. 2003a], seasonal exchange with the terrestrial biosphere [CASA model, Potter et al., 1993; Field et al., 1995; Randerson et al., 1997], and air-sea fluxes [gridded data base of Takahashi et al., 1999]. The variability of CO_2 concentrations in the TRACE-P flight region is determined by combustion sources and the terrestrial biosphere, with negligible contribution from the ocean. We do not discuss further the role of ocean exchange in this study.

The CASA fluxes employed here are driven by climatological Normalized Difference Vegetation Index (NDVI) products and mean temperature and precipitation distributions [Randerson et al.1997; Potter et al. 1993]. Fluxes derived for the year 2001 (J. Randerson, G. van der Werff, pers. comm.), yield similar values for the China region (5920 Gg C/day) as the climatology (5840 Gg C/day). These CASA fluxes do not resolve the diurnal variation of terrestrial biospheric CO_2 emissions. We evaluated the resulting error in our analysis of TRACE-P aircraft data by testing a prototype version of the CASA fluxes that accounts for diurnal variation [Olsen and Randerson, 2003]. The resulting effects were not significant, which can be explained by the small amplitude of the diurnal cycle in the CASA fluxes for temperate Asia in February-April, and by the 1-2 day oceanic fetch of the continental outflow sampled in TRACE-P [Fuelberg et al. 2003].

The CO_2 simulation was spun up from January 1, 2000 and run for 16 months to the end of April 2001. Due to the omission of interannual variability in terrestrial biospheric exchange and air-sea fluxes, modeled CO_2 concentrations display a small positive bias, relative to observations, at the beginning of the TRACE-P simulation (March 1st, 2001). The modeled distribution is, therefore, adjusted downwards, with a single global subtraction, to eliminate this bias and to match observations from the GLOBALVIEW-CO_2 [2002] database at the start of the TRACE-P period.

Our a priori CO simulation is that reported by Palmer et al. [2003a]. It includes the a priori Asian CO emission inventories for fuel combustion and biomass burning from Streets et al. [2003] and Heald et al. [2003a] respectively. Chemical sources of CO include oxidation of methane and NMHCS as described by Duncan et al. [2003b]. Loss of CO by oxidation is computed using archived 3-D OH concentration fields from a GEOS-CHEM tropospheric chemistry simulation [Martin et al. 2003]. We also consider, in section 6.3, the a posteriori CO simulation of Palmer et al. [2003a], where the source estimates have been adjusted using an inverse analysis of the TRACE-P CO measurements. The main changes relative to the a priori simulation are a 54% increase in anthropogenic Chinese emissions and a 74% decrease in biomass burning in Southeast Asia.

6. Interpretation of CO_2-CO correlations in TRACE-P

6.1 Simulations based on a priori inventories

Our analysis of CO_2-CO relationships in Asian outflow during TRACE-P focuses on boundary layer observations (> 840 hPa) in two regions (Figure 1). We choose to focus on the boundary layer data because they are most strongly influenced by Asian outflow and least affected by model transport errors [Kiley et al., 2003]. To compare model and observations we averaged the 1-minute merged data for TRACE-P CO_2 and CO measurements over the model grid resolution (2^ox$2.5^o$and 48 vertical levels). The model simulations were sampled along the flight tracks. Figures 6 and 7 present the mean vertical profiles of observed and simulated concentrations of CO_2 and CO. Good agreement is found for CO_2 in the free troposphere, implying no significant model bias in the simulation of the mid-latitude background. However, the CO_2 simulation is too high in the boundary layer by 1.9 ppmv for the outflow regions 1 and 2 (China and Japan). The model CO is too low, by 23 ppbv for the ensemble of data and by more in the boundary layer, due to an underestimate of anthropogenic sources in China [Palmer et al. 2003a].

Boundary layer CO_2/CO slopes from model and observations are shown in Figure 8. Mean CO_2/CO slopes were derived by linear regression using the reduced major axis method and are presented in Table 3 (as the 'A Priori' scenario). The mean observed values differ by more than a factor of 2 between Region 1 (18) and Region 2 (46). This is consistent with greater Japanese influence in region 2. The model slopes generated from a priori source inventories are too high, by almost a factor of 3 for region 1 and a factor of 2 for region 2, which reflects the model overestimate of CO_2 concentrations and the model underestimate of CO (Figures 6 and 7). We find that within the model world, the boundary layer CO_2/CO slopes are consistent with regional source ratios (e.g., in region 1, the model slope of 55 (Table 3) matches the a priori Chinese source ratio of 56 (Table 2)). We conclude that the discrepancy between modeled and observed boundary layer CO_2/CO slopes reflects errors in the a priori inventories as discussed below.

6.2 Constraints on CO_2 emissions using CO_2/CO slopes

In this section we combine the simulations of CO_2, CO and CO_2/CO slopes to obtain top-down constraints on Asian CO_2 emissions. We previously showed that the model with a priori source estimates overestimates CO_2 concentrations in the boundary layer with the largest discrepancy apparent in Chinese outflow. Reconciliation of modeled CO_2/CO slopes and mean CO_2 concentrations with the observations specifically requires a reduction in a regional CO_2 source with a CO_2/CO source ratio higher than the regional model mean value of 55. Among the Chinese CO_2 sources of Table 1, the only one that meets this criterion is the terrestrial biospheric flux. Decreases in net biospheric emissions of 45% for China (from 5840 to 3210 Gg C/day) and 10% for boreal Asia (defined as the Asian landmass east of 60^{o}E and north of 55^{o}N) give a better match between modeled and observed CO_2 in regions 1 and 2 and represent the 'Decreased Biosphere' scenario in Figure 6. These CO_2 emission changes eliminate the boundary layer overestimate of the CO_2 observations in all regions (Figure 6). They also decrease the modeled CO_2/CO slopes (Table 3) but not sufficiently to match the observed slopes. An increase in CO emissions is further required and will be discussed later.

The net biospheric flux in the model represents the balance between two large modeled fluxes defined by the CASA model (CO_2 uptake from NPP and emission from heterotrophic respiration) and is subject to some uncertainty. For the seasonal TRANSCOM inversions, Gurney et al. [2003] employ prior uncertainties on the CASA net biospheric fluxes in temperate Asia of 2.0, 2.2, and 2.5 Gt C/year for February, March and April, respectively (equivalent to 73%, 80%, and 92% of the net biospheric flux). Our 45% adjustment of the net biospheric source from China is within this range of uncertainty. However, matching the observed CO_2/CO slope of 18 in China outflow (Region 1, boundary layer), would require a decrease in Chinese biospheric emissions of almost 200% (i.e., China would have a net biospheric uptake of 5800 Gg C/day for the TRACE-P period). Such a change would be outside the estimated range of uncertainty on the a priori, and it would reduce the mean modeled CO_2 in the Region 1 boundary layer to 374.8 ppmv, a value much lower than observed (377.1 ppmv).

Since consistency between observed and modeled CO_2/CO slopes is not achievable through further reduction of CO_2 emissions, error in a priori emission estimates for CO, as previously identified by Carmichael et al. [2003b] and Palmer et al. [2003a], is a likely explanation. Streets et al. [2003] note a possible underestimate of biofuel or domestic coal emissions in their CO inventory due to under-reporting of domestic coal use in rural central China. Their stated uncertainty (95% confidence level) for Chinese fuel CO emissions (156%) is significantly larger than the corresponding uncertainty on Chinese fuel CO_2 emissions (16%).

Inverse modeling of the TRACE-P CO observations, using GEOS-CHEM as the forward model, calls for a 54% increase in Chinese fuel CO emissions relative to the Streets et al. [2003] a priori and a 74% decrease in Southeast Asian emissions relative to the Duncan et al. [2003a] a priori, with smaller changes for the other CO sources [Palmer et al. 2003a]. Figure 7 shows the resulting vertical profiles. There is still a low bias in the CO simulation, in part because the inversion assigns large transport errors to the simulation of urban plumes observed in the boundary layer outflow. Including the a posteriori CO emission estimates of Palmer et al. [2003a] in our analysis (new model scenario 'Decreased Biosphere/A Posteriori CO') further decreases modeled CO_2/CO slopes in the boundary layer to 32 in Region 1 and 47 in Region 2 (Table 3). While the modeled slope in Region 2 (47) is now close to the observed boundary layer value of 46, the slope in Region 1 (32) is still higher than the observed value of 18; this difference may result from the inability of the model to simulate observed urban plumes with high CO. We note that filtering the CO measurements to remove the influence of outliers as was done in the Palmer et al. [2003a] analysis, results in an observed boundary layer slope in Region 1 of 30 mol/mol (Figure 8), yielding a better match between observations and the Decreased Biosphere/A Posteriori CO' scenario.

6.3 Implications of the a posteriori CO simulation for Asian CO_2 emissions

The large adjustments to the Chinese and biomass burning CO sources in the TRACE-P inverse model analysis of Palmer et al. [2003] raise the question as to the implications for CO_2. The bottom-up combustion inventories for CO and CO_2 were constructed by applying species-specific emission factors to common databases of activity rates (and abatement technologies when relevant). The errors in the a priori inventories for CO could be due to errors in activity rates, which would have associated implications for CO_2 emissions, or in CO emission factors, which would not.

We first examine the Chinese anthropogenic sector, for which Palmer et al. [2003] adjusted CO emission upward by 54% relative to the a priori of Streets et al. [2003]. Uncertainties given by Streets et al. [2003] on their Chinese fuel inventories are 156% for CO and 16% for CO_2, and Table 4 presents their partitioning of Chinese fuel emissions among the main sectors. The CO_2/CO emissions ratios, aggregated for each sector, vary from 5 mol/mol (transport) to 38 mol/mol (industry). Streets et al. [2003] assumed western emissions controls standards for the power plants included in their analysis and hence estimate negligible CO emissions from this sector.

Likely candidates for the underestimate in CO emissions are the domestic coal and biofuel sectors [Carmichael et al. 2003b], and small heavily polluting power plants unaccounted for in official industry statistics [Bradsher, New York Times, 2003]. We consider first the possibility of an underestimate in the domestic fuel sector activity with two extreme scenarios, 'Increased Domestic Coal' and 'Increased Biofuel', for which the CO underestimate derives entirely from these sub-sectors. Our analysis approach here is to estimate the changes in sector activity based on the projected increase in CO emissions from Palmer et al. [2003a], and then derive consistent CO_2 emissions changes. The implied increases in activity rates are a factor of 7.5 for the domestic coal sector (equivalent to CO_2 emissions increasing from 181 Gg C/day to 1370 Gg C/day), or 2.5 for the biofuels sector (equivalent to CO_2 emissions increasing from 556 Gg C/day to 1396 Gg C/day). These changes in CO_2 (32% to 45% of Chinese anthropogenic emissions) fall outside the 16% uncertainty estimated by Streets et al. [2003]

Implementation of these CO_2 emissions in the GEOS-CHEM model leads to a mean overestimate of CO_2 concentration in the boundary layer of Region 1 by 1.2 ppm ('Increased Biofuel') and 1.3 ppm ('Increased Domestic Coal'). The corresponding mean CO_2/CO slopes are 35 mol/mol and 37 mol/mol which degrade the simulation of observations relative to the 'Decreased Biosphere/A Posteriori CO' scenario of section 6.2. Reconciliation of simulated boundary layer CO_2 with the observations, for the 'Increased Biofuel' scenario, for example, would require further decreases in the Chinese biospheric flux (of 1200 Gg C/day). The overall required decrease in the Chinese biospheric flux would thus be 3800 Gg C/day (also accounting for the contribution from the 'Decreased Biosphere' scenario) and equivalent to 65% of net Chinese biospheric emissions. We note, however, that such an adjustment would still be within the uncertainty bounds on the springtime CASA fluxes from Gurney et al. [2003].

We next consider the possibility of errors in CO emission factors, rather than sector activity rates, as being responsible for the underestimate of Chinese CO fuel emissions. Under this assumption, attribution of the CO emissions adjustment implied by Palmer et al. [2003] to the domestic fuel sub-sectors would result in unrealistically high CO emission factors, 15% CO for biofuels or 29% for domestic coal.

Unaccounted CO emissions from older heavily polluting power plants, designated for closure but apparently still active [Bradsher, New York Times, 2003], is another possibility. While modern power plants are equipped with emissions controls designed to minimize CO emissions, older plants lacking this technology, may provide a significant source of CO. Streets et al. [2003] do not account for such plants in their analysis. If the underestimate of Chinese anthropogenic CO emissions were due solely to this source then the overall CO emission factor for the power plant sector (assuming no increase in CO_2 emission) would be 8%, which again seems unrealistically high.

It appears therefore that no single factor can easily explain the underestimate of anthropogenic Chinese CO emissions in the Streets et al. [2003] inventory. A multiple explanation, involving a combination of underreporting of sector activity together with low bias in CO emission factors, seems most likely. The former would imply a reduction in the biospheric source of CO_2 beyond that used in our 'Decreased Biosphere/A Posteriori CO' but still within the uncertainty range estimate of Gurney et al. [2002] for the springtime CASA fluxes.

We now examine the consequences of the large decrease in biomass burning CO emissions (-74% relative to the a priori in Southeast Asia) estimated by Palmer et al. [2003a]. If this were due to an overestimate of biomass burning activity (as opposed to an overestimate in emission factor) then CO_2 emissions should correspondingly decrease. We focus on the free troposphere north of 30^o N, where biomass burning signals during TRACE-P were strongest [Carmichael et al. 2003a; Liu et al. 2003; Li et al. 2003]. Reducing Southeast Asian biomass burning emissions by 74% worsens the modeled CO_2 underestimate in the free troposphere by 0.15 ppm in comparison to the 'Decreased Biosphere/A Posteriori CO' simulation (model bias changes from –0.5 to –0.65 ppm). The effect on mean modeled CO_2/CO in this region is negligible (decrease of less than 2 mol/mol). These relatively small effects yield inconclusive results on the implied biomass burning decrease within our analysis framework. A formal coupled inversion of CO_2 and CO may better identify which scenarios are robustly constrained by the measurements and a priori uncertainties. For now, our best case scenario remains the 'Decreased Biosphere/A Posteriori CO' of section 6.2.

The results from our best case scenario of East Asian CO_2 emissions can be compared to those of Vay et al. [2003], who used their TRACE-P CO_2 measurements to estimate CO_2 export fluxes from Asia for March-April, 2001. They calculated export fluxes as the product of gridded zonal wind vectors and CO_2 enhancements over a background value for the NW Pacific (20^o-40^{o}N, 120^o-150^o E, and 0-12 km). Two separate fluxes were calculated; (a) relative to a fixed background concentration and (b) relative to a latitudinally varying background. Vay et al. [2003] argue that using a fixed background yields a total CO_2 export from the Asian continent (13.93 Tg C /day), while using a latitudinally varying background yields the anthropogenic component of that export (6.37 Tg C/day). They conclude that 54% of the total east Asian CO_2 outflow is from biospheric sources. In comparison, our 'Decreased Biosphere/A Posteriori CO' scenario yields total CO_2 emissions for the East Asian region (as defined in Figure 3) of 13.1 Tg C/day with a biospheric contribution of 40%. The difference between the two estimates is not large, and we note that use of the Streets et al. [2003] estimates of East Asian anthropogenic emissions (7.4 Tg C per day), instead of Vay et al.'s own estimate (6.4), yields a biospheric contribution of 47% in their calculation, which is closer to our estimate of 40%.

7. Summary

We have used observed CO_2/CO correlations in Asian outflow from the TRACE-P aircraft campaign (March-April 2001) to constrain different regional components of the East Asian CO_2 budget. The combustion sources of CO_2 and CO display characteristic emission ratios ranging from 12 mol/mol for biomass burning to over 100 for Japanese fossil fuel combustion. The biospheric source of CO_2 is almost independent of CO as the biogenic source of CO is small. These signatures yield regionally distinct emission ratios which reflect the relative proportions of the various sources within each region.

Examination of CO_2/CO correlations from individual TRACE-P flights indicates indeed distinct regional signatures. Outflow from northeast China has a low CO_2/CO slope (10 - 20 mol/mol), reflecting inefficient combustion sources from domestic coal and biofuel. Measurements over Japan indicate much higher slopes (60 - 80 mol/mol) reflecting CO emissions controls in that country.

We used a global 3-D simulation of CO_2 and CO from the GEOS-CHEM model to interpret the observed CO_2/CO slopes in terms of specific sources. When driven by our best a priori estimates of sources (the Streets et al. [2003] anthropogenic inventory, the Duncan et al. [2003a] biomass burning inventory, and the CASA biospheric model), the model overestimates both CO_2 concentrations and CO_2/CO slopes in the Chinese outflow. This implies that the regional springtime biospheric source of CO_2 from the CASA model is too high. Reconciliation of model CO_2 concentrations with the TRACE-P observations is achieved by decreasing net biospheric emissions in China by 45% and in boreal Asia by 10%, an adjustment which is within the range of uncertainty of the CASA model fluxes. This change alone, however, is insufficient to reconcile the modeled boundary layer CO_2/CO slopes with observations. Previous analyses of the TRACE-P CO data had found that Chinese anthropogenic CO emissions must be higher than in the Streets et al. [2003] inventory (by 54% according to formal inverse model analysis by Palmer et al. [2003a]). We find that such an adjustment improves the simulation of the CO_2/CO slopes. The analysis of Palmer et al. [2003a] also calls for a 74% decrease of biomass burning CO emissions from Southeast Asia relative to the a priori inventory of Duncan et al. [2003a], but the corresponding signal in CO_2 along the TRACE-P aircraft flight tracks is too weak to provide independent information on this issue.

Analysis of our CO_2 simulation indicates that a combination of factors, including underreporting of sector activity (domestic and industrial combustion) together with low bias in CO emission factors, is likely responsible for the underestimate of Chinese anthropogenic CO emissions in the Streets et al. [2003] inventory. Underreporting of sector activity would imply increases in Chinese anthropogenic CO_2 emissions and would require further decrease of the Chinese biospheric CO_2 source to reconcile simulated and observed concentrations of CO_2 in the Asian outflow. Such an adjustment would still be within the uncertainty bounds on the springtime CASA fluxes. An optimal set of emissions adjustments to match the combined constraints from observed CO_2 and CO concentrations as well as CO_2:CO correlations will require a more formal inverse analysis.

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Table 1: A priori East Asian CO_2 and CO emissions for the TRACE-P period (Gg C/day)[1]

	CHINA		JAPAN		KOREA		INDIA		SOUTHEAST ASIA	
	CO_2	**CO**	**CO_2**	**CO**	**CO_2**	**CO**	**CO_2**	**CO**	**CO_2**	**CO**
Fossil Fuel[2]	1930	75	964	8.2	380	5.8	484	20	686	19
Biofuel[2]	640	53	21	2.3	31	4.7	496	34	314	45
Biomass[2] Burning	270	22	11	0.9	4	0.3	552	46	1160	96
Biosphere [3]	5840	4	411	0.5	156	0.1	1080	11	328	31
TOTALS	8680	154	1410	11.9	571	10.9	2610	111	2490	191

[1] A priori estimates of CO_2 and CO fluxes for the regions of Figure 3. Values are averages for the TRACE-P period (March 1 – April 6, 2001).

[2] The combustion sources of CO include a small contribution from oxidation of anthropogenic NMHCs, computed as described by Duncan et al. [2003b]

[3] The biospheric flux term for CO_2 represents net emission or the negative of Net Ecosystem Production (NEP), where NEP = Net Primary Production (NPP) – Emissions from heterotrophic respiration (R_h), as defined by the CASA model [Randerson et al. 1997]. The biospheric source of CO represents production from oxidation of biogenic NMHCs as described in Duncan et al. [2003b]

Table 2 : A priori East Asian CO_2/CO molar emission ratios.[1]

	CHINA	JAPAN	KOREA	INDIA	SOUTHEAST ASIA
Fossil Fuel	26	117	65	24	36
Biofuel	12	9.1	6.6	15	7.0
Biomass burning	12	12	12	12	12
Biosphere[2]	1460	822	1560	98	10.6
Regional Average	56	119	52	24	13

[1] Derived from the a priori inventories of Table 1 for the period March – April 6, 2001.
[2] A combination of high biogenic NMHC emissions and small NEP CO_2 fluxes explains the low biospheric CO_2/CO emissions ratio for Southeast Asia.

Table 3: CO_2:CO correlation slopes (mol/mol) and R^2 along the TRACE-P flight tracks[1]

	OBSERVATIONS[2]	MODEL		
REGION		A Priori	Decreased Biosphere	Decreased Biosphere/ A Posteriori CO
Region 1:China outflow (> 840 hPa)				
SLOPE	18 (30)	55	46	32
R^2	*0.52*	*0.66*	*0.65*	*0.67*
Region 2:Japan outflow (> 840 hPa)				
SLOPE	46 (49)	80	69	47
R^2	*0.4*	*0.73*	*0.72*	*0.64*

[1] The comparisons are for the regions of Figure 1. The mean CO_2/CO slopes are derived by linear regression using the reduced major axis method on each set of regional data. The 'A Priori' model simulation uses the CO_2 and CO inventories of Table 1. The 'Decreased Biosphere' simulation imposes a reduction of net biospheric CO_2 emissions (45% in China and 10% in boreal Asia). The 'Decreased Biosphere/A Posteriori CO' simulation includes in addition, the a posteriori CO emissions estimates of Palmer et al. [2003a], and achieves the closest agreement to the observed CO_2/CO slopes.

[2] Values in parentheses represent the mean CO2/CO slopes obtained when CO observations are filtered to remove outliers as was done in the Palmer et al. [2003a] analysis

Table 4 : Chinese fuel CO_2 and CO Emissions by sector in the Streets et al. [2003] inventory [1]

	Biofuel	**Domestic Coal**	**Industry**	**Transport**	**Power[2]**	**Cement**
CO_2 (Gg C/day)	556	181	809	228	710	156
CO (Gg C/day)	42	9.6	21	45	-	-
CO_2/CO (mol/mol)	13	19	38	5.1		

[1] Small differences between these totals and those of Table 1 are due to different regional definitions for China. The numbers here are computed for the Chinese region defined in Streets et al. [2003]. The fluxes of Table 1 are computed for the regions of Figure 3.
[2] Streets et al. [2003] assumed western emissions controls standards for the power plants included in their analysis and hence estimate negligible CO emissions from this sector.

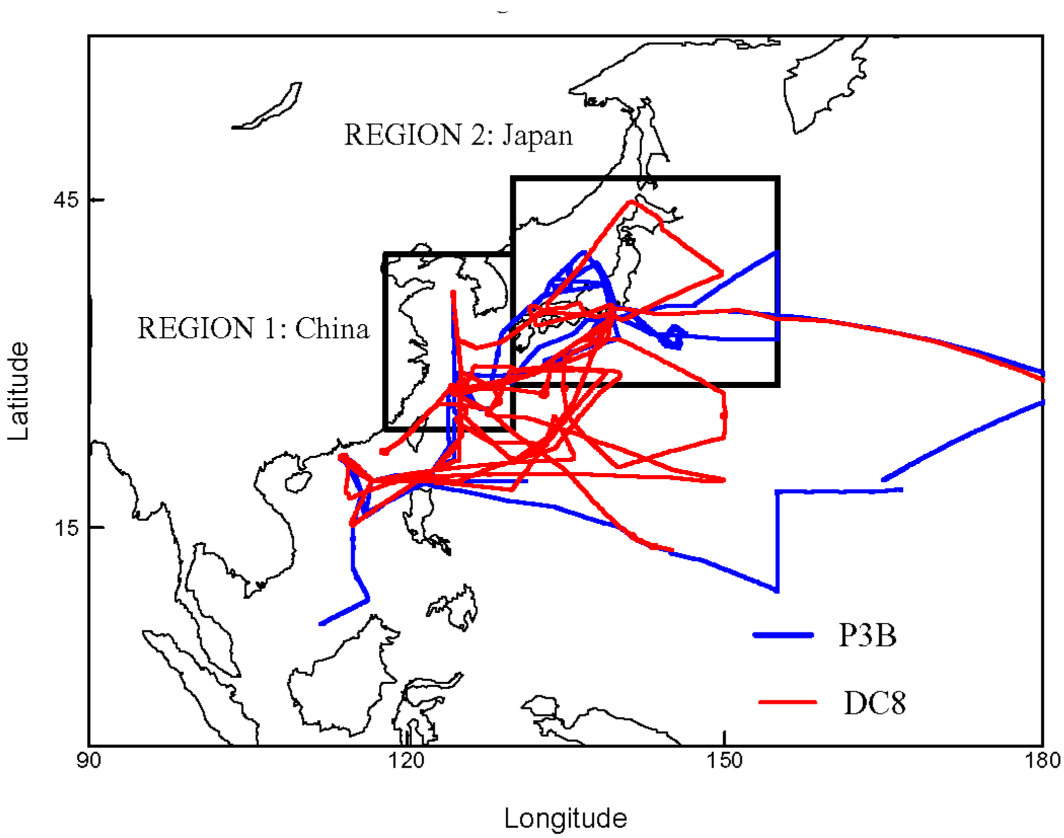

Figure 1 : TRACE-P flight tracks off the Asian coast. Also shown are the two regions over which flight data were aggregated for comparisons with model simulations (see section 6.1)

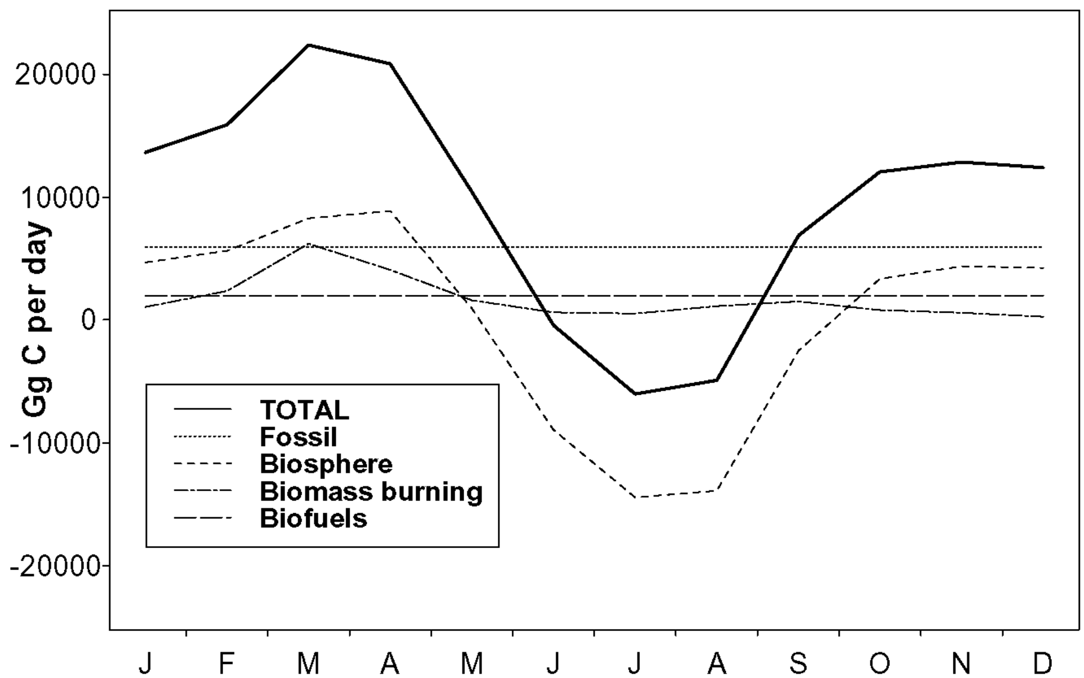

Figure 2 : Seasonal variation of East Asian CO_2 surface fluxes derived from the a priori inventories of section 3. East Asia is defined here as the continental domain covered by the five regions of Figure 3.

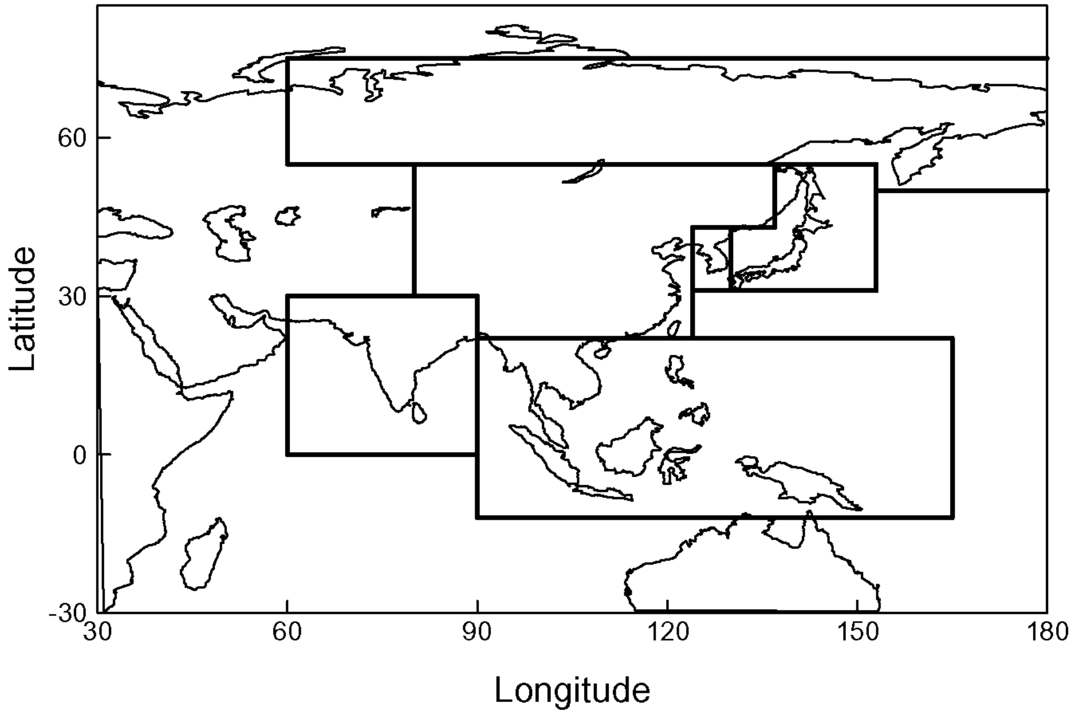

Figure 3 : Regions used in the emission inventory analysis: China, Japan, Korea, India, Southeast Asia and boreal Asia.

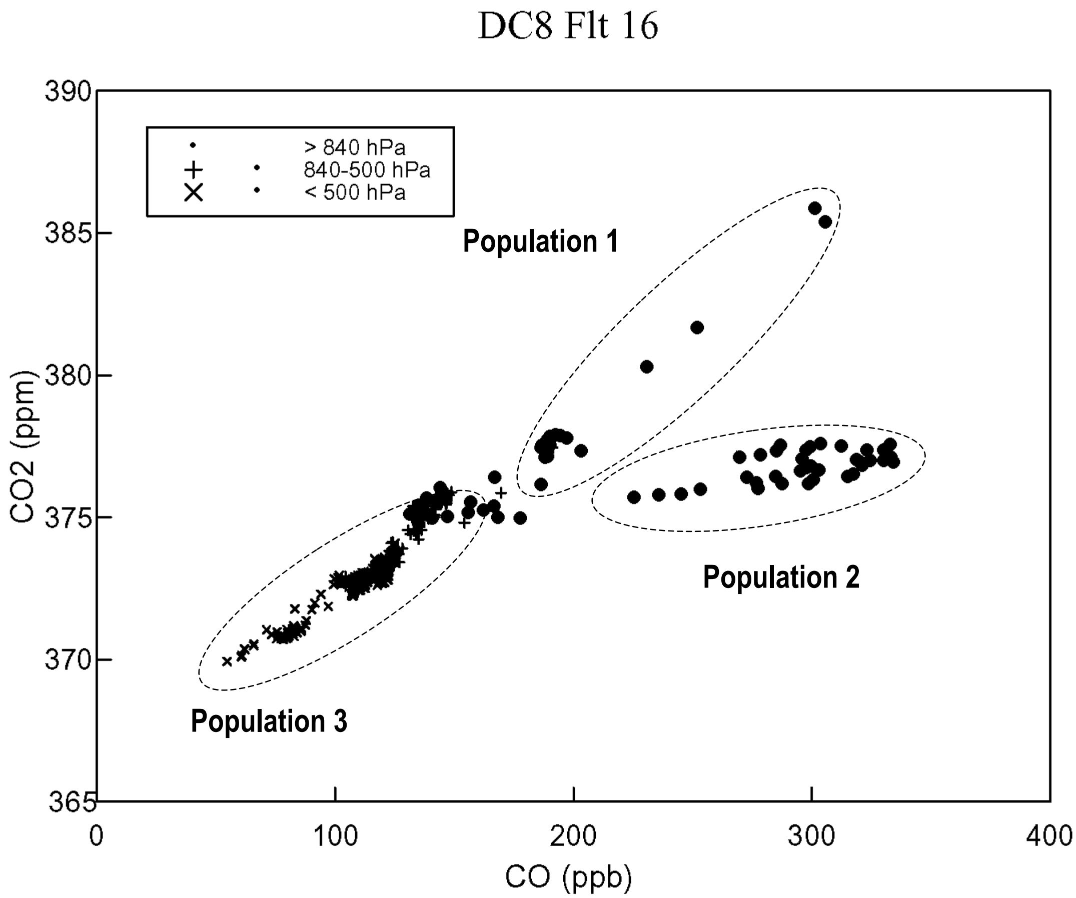

Figure 4 : CO2/CO correlations measured on DC8 Flight 16 out of Yokota Air Force Base, Japan (March 29, 2001). Mean slopes for the different populations are 65 for Population 1 (boundary layer ascent out of Japan), 12 for Population 2 (boundary layer outflow from China), and 60 for Population 3 (mid-tropospheric background).

P3B : Flight 14

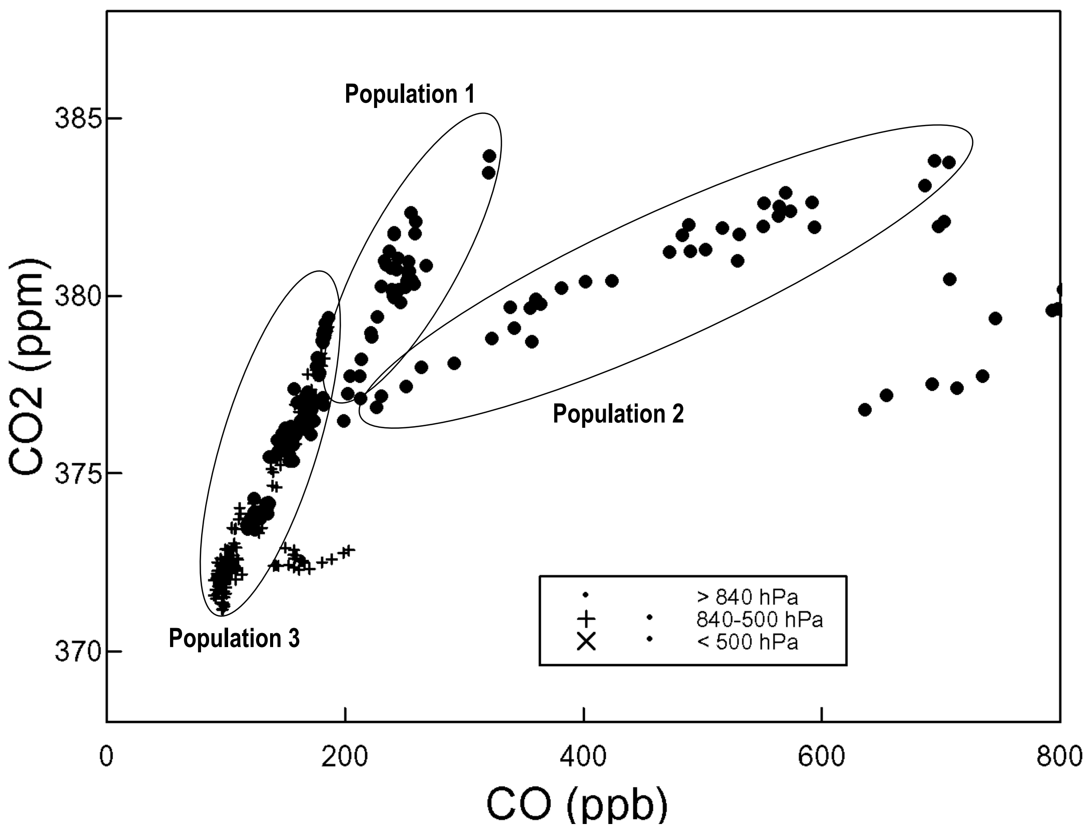

Figure 5 : CO2/CO correlations measured on P3B Flight 14 over the Yellow Sea (March 18, 2001). Mean slopes for the different populations are 75 for Population 1 (mixed boundary layer outflow from China, Korea and Japan), 22 for Population 2 (boundary layer outflow from north-east China) and 80 for Population 3 (mid-tropospheric background air). The measurements characterized by high CO (> 600 ppbv) that are not included in Population 2 are from the Beijing urban plume.

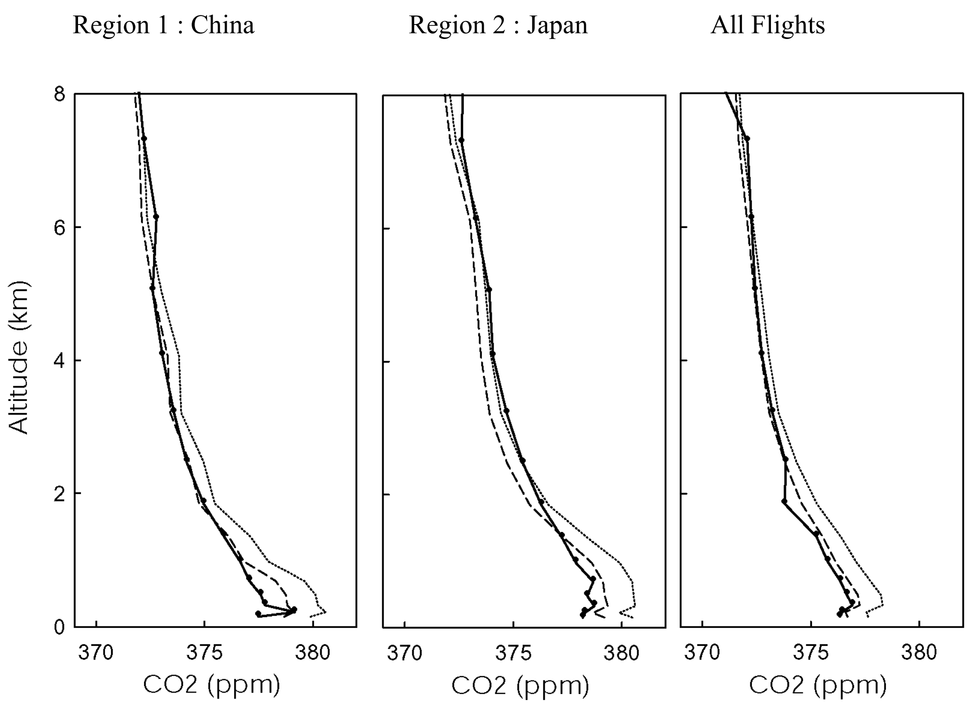

Figure 6 : Mean variation of CO2 concentrations with altitude along the TRACE-P flight tracks, for Regions 1 and 2 and for the ensemble of flights west of 150^o E. The aircraft observations (bold line) are compared to GEOS-CHEM model results using the 'A Priori' emissions inventories (dotted line) and the optimized 'Decreased Biosphere' emission scenario of section 6.2 (dashed line).

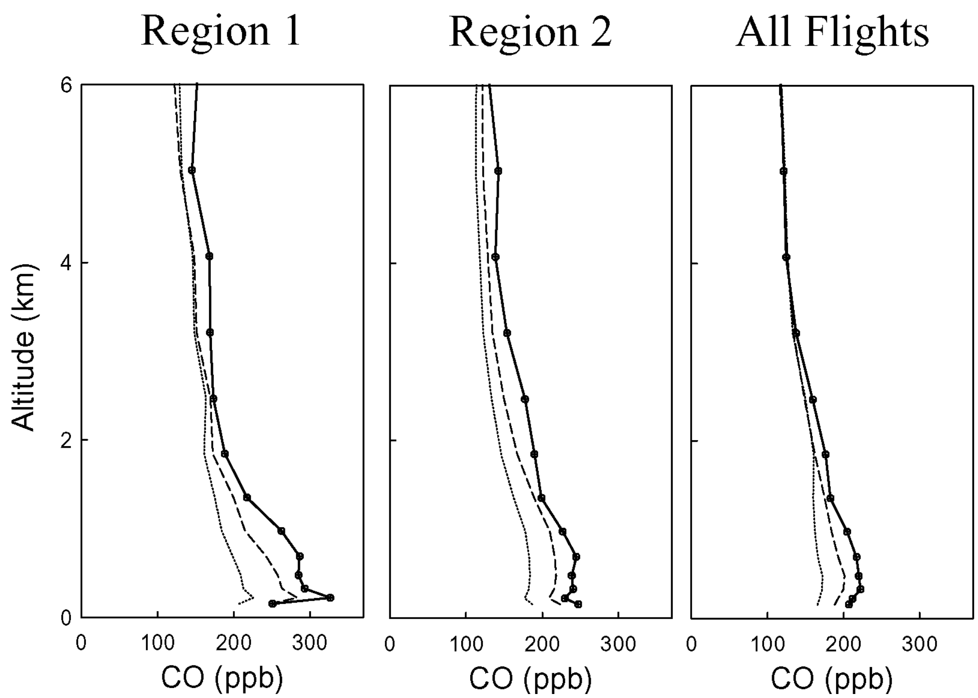

Figure 7 : Mean variation of CO concentrations with altitude along the TRACE-P flight tracks, for Regions 1 and 2 and for the ensemble of flights west of 150^o E. The aircraft observations (bold line) are compared to GEOS-CHEM model results using the 'A Priori' emissions inventories (dotted line) and the optimized a posteriori estimates of Palmer et al. [2003a] (dashed line).

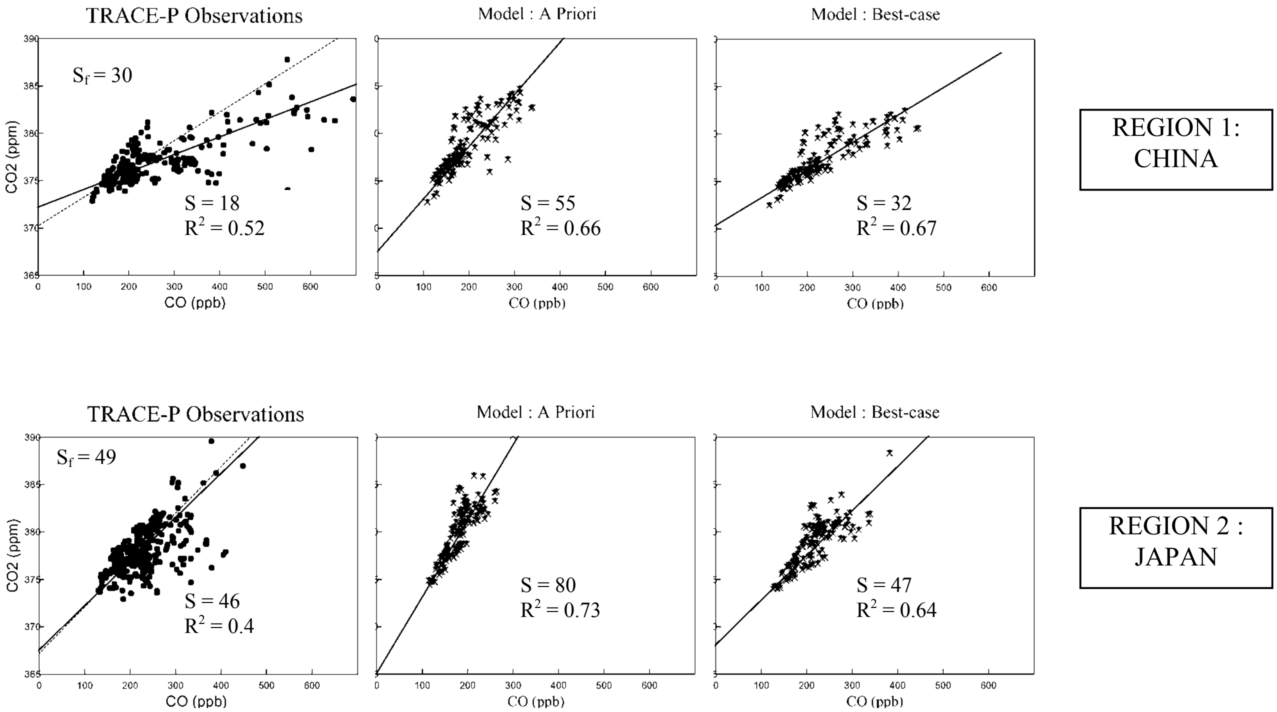

Figure 8 : Comparison of boundary layer (> 840 hPa) CO_2:CO correlations in Regions 1 and 2 from (a) TRACE-P observations (left panels) and GEOS-CHEM simulations for (b) A priori inventories (middle panels), and (c) our Best-case scenario ‘Decreased Biosphere/A Posteriori CO’ (right panels). The model is sampled along the aircraft flight tracks. The RMA linear regression with slope S is plotted (solid line). Also shown is the RMA regression slope, S_f, (dotted line) when the TRACE-P CO observations are filtered to remove outliers, as in the analysis of Palmer et al. [2003].